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Temperature-dependent O1s NEXAFS of Lightly Doped Ca_2RuO_4

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Introduction: Ca_2RuO_4, a layered perovskite related to the superconductor Sr_2RuO_4 and with RuO_6 octahedra as its structural backbone, undergoes insulator-metal transitions upon doping with Sr and with rare earth (RE) elements like La and Nd. In the former case the transition can be traced to a reduced role of correlation effects, which is related to doping-induced structural rearrangements in the RuO_6 octahedra that in turn lead to an increase in valence band width. In the latter case, the extra electrons injected by doping ("band filling") may have an additional effect on the transition. In all cases, doping levels below 10% are sufficient to induce metallic behavior at room temperature and to shift the transition temperature T_{MI} to the 200-250 K range [1]. While composition-dependent information on the electronic structure of these compounds is available [2], similar studies on how varying the temperature through T_{MI} affects the electronic structure for a given composition have only recently been started for Sr-doped compounds [3]. The present work expands on this by concentrating on doping by RE elements.

Methods and Materials: X-ray absorption (NEXAFS) measurements were performed at the O 1s edge, detecting the unoccupied density of states with O2p character. Bulk-sensitive fluorescence detection mode was employed, and the sample temperature could be varied between 78 K and 380 K. High-quality single crystals of Ca_2RuO_4 doped with Sr, La, Nd, and Sn at various levels in the "few percent" range were available, whose size allowed polarization-dependent measurements that can distinguish between in-plane and out-of-plane character of the O2p orbitals involved. Considering possible hybridizations with Ru4d orbitals an assignment of spectral contributions to the inequivalent O sites in the RuO_6 octahedra (apical and planar) is possible.

Results and Conclusions: Upon variation of temperature, the in-plane spectra **E**||(*a*,*b*) for all RE-doped materials exhibit a clear and systematic redistribution of spectral weight between the two spectral features closest to onset: a peak "A" related to the apical O sites, and a peak "P" derived from the planar sites, both π-hybridized with the t_{2g} subset of Ru4d orbitals. Peak A dominates in the insulating phase, peak P in the metallic phase, which corresponds well with the observation that conductivity is mainly carried in the planes. A hysteresis effect in the peak height ratio P/A when going through T_{MI}, like the one observed in [3] for a Sr-doped compound, is much less obvious here. This might be an illustration of a basic difference between Sr doping and RE doping, and deserves further experimental attention.

The temperature effect for **E**||*c* is smaller and is dominated by reduced phonon broadening with lower temperature. Some changes may be present in the position and weight of a weak unoccupied structure identified in [2] to be an upper Hubbard band, and would further illustrate the delicate interplay between band filling, correlation, and structure that drives and characterizes this series of insulator-metal transitions.

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References: [1] see, e.g., G. Cao et al., PRB 61, 5053 (2000). [2] S. Gerhold et al., to be published (2002). [3] T. Mizokawa et al., to be published (2002).